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Issues on nitrogen oxides concentration reduction in the combustion products of natural gas

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Abstract

The issues on nitrogen oxides formation during the fossil fuels combustion in the furnace-tube boiler have been considered, taking into consideration the national and foreign literature of recent years. Chemical reactions as well as mathematical models describing the emergence and further changes in the concentrations of thermal, fast NO_x are proposed. The coefficient impact assessment of the furnace-tube boiler finning on the composition of gaseous fuels combustion products is provided.

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Keywords: burning; nitrogen oxides; thermal; fast; fuel; boiler

1. Introduction

The largest atmosphere polluters are the combustion products of thermal electric power plant and boiler-house plant [1, 2]. In burning the fossil fuels a large number of toxic substances such as nitrogen oxides and sulfur, ash, etc. gets into the atmosphere along with the boiler combustion gases. Currently, the natural gas and fuel oil share in the Russian heat power industry is more than 73% of the total combustible fossil fuels.

The main toxic component produced in the combustion of natural gas and fuel oil in the furnaces of the steam and water-heating boiler are nitrogen oxides NOx. Nitrogen oxides have a negative impact on human health, particularly on the respiratory system. Although the amount of nitrogen oxides produced naturally is much higher than the emissions from the human activity, it must be considered that the anthropogenic emissions of nitrogen

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oxides are concentrated in the areas of anthropogenic activities. Therefore, the concentration of NOx in urban areas is higher than the natural background concentration.

Conceptually there are different approaches of reducing the harmful substances emissions with the combustion gases into the atmosphere. They can be divided into the following three groups [1-5]: harmful components removal from the fuel by its complex processing before combustion in the boiler; direct impact on the harmful substances formation mechanism during the combustion of natural raw fuel in the combustion chamber; the combustion products (the combustion gases) scrubbing from harmful compounds.

The aim of this paper is to study computationally the combustion of gaseous fuel in the furnaces-tube boilers with the simultaneous limitation of nitrogen oxides emissions. These processes are poorly understood with respect to boilers of low power and are of some interest in the development of modern autonomous heating sources.

2. Nitrogen oxides formation modelling

To describe the turbulent flow of the reacting gases in the furnace-tube boiler the model of turbulence with two equations is used. This model of turbulence is called *k-ε* with two equations (k is the turbulent kinetic energy; ε is the value of kinetic energy dissipation).

In burning the fossil fuels in boilers the nitrogen of the air, interacting with oxygen, forms oxides:

$NO_x = NO + NO_2 + N_2O$.

The main share of NO_x (95-99 %) generated in the combustion products of steam and water-heating boilers is carbon monoxide NO. Dioxides NO2, nitrous oxide N_2O are produced in much smaller quantities.

Nitrogen monoxide formation in the CH_4 fossil fuels combustion takes place due to the oxidation of N_2 atmospheric nitrogen. At present there are three mechanisms of the nitrogen oxides formation: thermal, fast and fuel. In the thermal and fast NOx formation a source of nitrogen is the air, and in the case of NO fuel formation the nitrogen components are the fuel.

The mechanism of thermal nitric oxide formation is proposed by Zeldovich and it includes the following reactions:

$O + N_2 \rightarrow NO + N$,

$N + O_2 \rightarrow NO + O$.

It was later supplemented by the reaction of atomic nitrogen with hydroxyl and was named as the extended mechanism of Zeldovich:

$OH + N \rightarrow NO + H$.

Thermal NOx formation reactions are characterized by high energy activation, so the formation of nitrogen oxides occurs at high temperatures exceeding 1800 K. Thermal NOx concentration rapidly increases from the beginning of the combustion zone and reaches the maximum value immediately after the maximum temperature zone. Further, the concentration of nitrogen oxides on the flame level remains practically unchanged. The expressions for the rate constants k of each of the three reactions are presented in [2] and are as follows:

$$k_1 = (1{,}8 \cdot 10^{11}) exp\left(-\frac{38370}{T}\right),$$

$$k_2 = (6{,}4 \cdot 10^{9}) exp\left(-\frac{3162}{T}\right),$$

$$k_3 = 3{,}0 \cdot 10^{13}.$$

The thermal NO formation defines the following main factors: the temperature in the combustion zone, the excess air ratio and the residence time of the combustion products in high temperature zone. The amount of the $S_{NO,thermal}$ component is determined by:

$$S_{NO,therma} = W_{NO} k_{thermal} [O][N_2],$$

where $k_{thermal}$=$2k_1$, W_{NO} is the molar mass of thermal NO; [O], [N_2] are molar concentration of oxygen and nitrogen.

Investigations on hydrocarbon fuels burning carried out by Fennimore [2, 5] showed that in the flame front the nitrogen oxides formation occurs in a very short time by a mechanism different to the one proposed by Zeldovich. The discovered nitric oxide was named as fast due to the high speed of its formation in the flame root. In the combustion zone proximity there were considerable amounts of hydrogen cyanide HCN, which could be explained by the molecular nitrogen reacting with hydrocarbon residuals:

$CH + N_2 \rightarrow HCN + N$,

$HCN + O_2 \rightarrow NO + \ldots$.

Fast oxides NO formation reaction occurs quite actively at 1200 - 1600 K temperature, when there is practically no formation of thermal nitric oxide.

The amount of the $S_{\mathrm{NO},\,prompt}$ component is defined by the expression [2]:

$$S_{\mathrm{NO},prompt} = W_{\mathrm{NO}} k_{prompt} [\mathrm{O_2}]^{1/2} [\mathrm{N_2}][Fuel] \left(\frac{W}{\rho}\right)^{\frac{3}{2}},$$

$$k_{prompt} = A_{prompt} \exp\left(-TA_{prompt}\right)/T,$$

where W_{NO} is the molar mass of NO, A_{prompt} is the Arrhenius number. The formulas for the reaction rates calculation are applicable only to flows with small values of Re (Reynolds number). In the turbulent fluctuations systems the parameters can have a dominant influence on the rate of NO formation. In this case statistical methods are used for calculation.

3. Computational techniques

Modeling of turbulent combustion processes with nitrogen oxides formation occurring in the furnace-tube boiler is carried out using ANSYS CFX program [2, 3].

The subject of the computational research is the cylindrical furnace-tube boiler (Fig. 1) and cylindrical furnace with internal finning (Fig. 2). Geometrical characteristics of the furnace are D = 0,5 m, L = 1,0 m, d_{in} = 0,08 m, d_{out} = 0,23 m.

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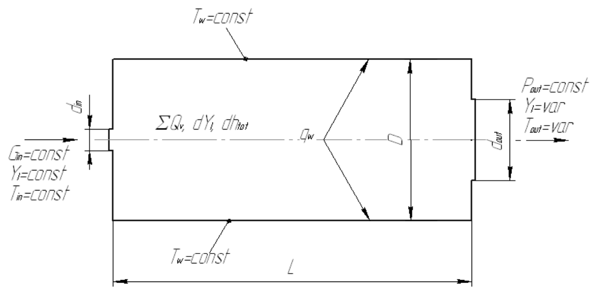

Fig. 1. Computational model of the furnace-tube boiler

In the no-flow boundary (the wall of the combustion chamber) the conditions of impermeability; pressure gradient equal to zero in the direction to the surface normal; constant wall temperature T_W are defined.

On the permeable border (the furnace entrance) flow of gaseous products G_{in}; the component Y_I concentration; T_{in}; mixture temperature; enthalpy change dh_{tot}; turbulence intensity, which is an approximate value for the internal flow in a pipe, are defined; the kinetic energy and the turbulence dissipation at the entrance are calculated. On the permeable border (the furnace exit) the P_{out} pressure, which is a characteristic feature of the system for combustion products removal, are defined. Reacting mixture is: the fuel is methane, the oxidizer is air.

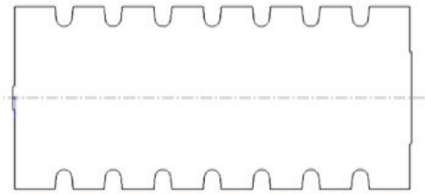

Fig. 2. Finned furnace scheme.

4. Results and discussion

It is found that there is an increase in convective phenomena with the use of furnace inside surface finning. It is accompanied with a decrease in the relative concentration of nitrogen oxides NO^*_x / NO_x (NO^*_x is the mass concentration of nitrogen oxides at the the furnace exit with the finning inside surface, NO_x is the mass concentration of nitrogen oxides at the furnace exit with the non-finning inside surface) at the exit from the furnace-tube boiler (Fig. 3), with a decrease of the maximum and average temperature *T* (Fig. 4) of the gas mixture inside the furnace and recycling processes in accordance with the equation:

$$\mathrm{NO} + \nu_F Fuel \rightarrow \frac{1}{2}\mathrm{N}_2 + \nu_{\mathrm{CO}_2}\mathrm{CO}_2 + \nu_{\mathrm{H}_2\mathrm{O}}\mathrm{H}_2\mathrm{O},$$

where v is the stoichiometric ratio; *Fuel* is some kind of fuel.

The law of the change NO^*_x / NO_x repeats the law of temperature change. When changing the finning ratio from 1.0 to 1.3 the convection heat transfer increases and the temperature decreases in volume. Thereafter, heat transfer is reduced due to the aerodynamic drag increase and the formation of dead zones between the fins.

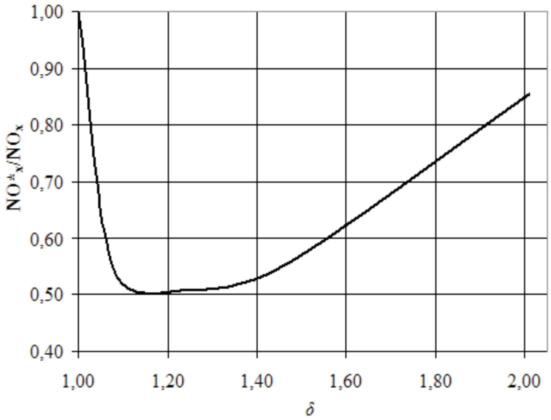

Fig. 3. Dependence of NO^*_x / NO_x relative consentration on the finning ratio.

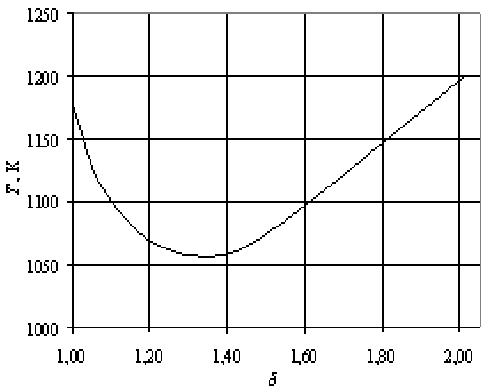

Fig. 4. Dependence of the volume-averaged gas mixture temperature on the finning ratio.

5. Conclusion

It is shown that the processes of combustion in the furnaces-tube boilers are always accompanied by the movement of gas such as air, gaseous fuel and combustion products and they are a set of aerodynamic, thermal and chemical processes. Also, in burning of the fossil fuels nitric oxide is formed due to the oxidation of nitrogen in the air.

Inside surface finning of the cylindrical furnace leads to:

1. Intensification of convective phenomena,
2. Increase of the combustion products recirculation and the gas temperature reduction;
3. Reduce of the nitrogen oxides concentration at the furnace exit.

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